Theory of Orbital Nematicity in Underdoped Iron Arsenides

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Recent finding of an *unusual* in-plane resistivity anisotropy in the underdoped 122-family at high temperature (T) suggests an orbital nematic (ON) order, posing a challenge to extant theories. The *sign* of the anisotropy contradicts expectations from weakly correlated as well as pure spin-only nematic views. Here, we show how such an ON order with accompanying structural distortion arises from *residual*, intersite and inter-orbital two-body interactions in an incoherent "bad metal" close to Mottness. Enhancement of orbital-selective incoherence is shown to be necessary for understanding transport anisotropy. Our results suggest that ON order, with subsequent antiferromagnetic order might be the *primary* competitor to superconductivity in Fe arsenides.

The "Iron Age" of high temperature superconductivity (HTSC) appears to reinforce the paradigm shift set in motion by HTSC in cuprates more than twenty years ago. Examining analogies and differences between cuprates and Fe-pnictides (FePn) can shed more light on both. While parent cuprates are antiferromagnetic (AF) Mott insulators, parent FePn are multi-band AF metals, with the Fermi surface (FS) constituted of several small pockets. Notwithstanding this difference, the normal metallic state (by chemical doping) in both defies even a qualitative description within traditional Landau Fermi Liquid (LFL) concepts. FePn thus appear to be anomalous metals giving way to unconventional superconductivity at high temperature (T) [1].

AF in FePn is universally preceded by, or coincides with, a tetragonal-to-orthorhombic (T-O) structural distortion. Recent work on 122- [2] and 1111-based [3] FePn reveals signs of enhanced nematic (N) susceptibility (χ_N) and *electronic* crystallinity upon underdoping, deepening the analogy with cuprates. Transport data puts strong constraints on theories purporting to describe the N-phase. In particular [2], (i) inspite of the fact that the N order leads to $b < a$ (here, a, b are planar unit cell distances), and (ii) the $\mathbf{q} = (\pi, 0)$ AF implies antiferromagnetically aligned spins along a and ferromagnetically aligned spins along b, the resistivity $\rho_b(T)$ along b *exceeds* $\rho_a(T)$ along a, an inexplicable result in a weak-coupling LFL [4] or pure spin-fluctuation [5] scenarios. In addition, $\rho_b(T)$ shows insulator-like behavior above T_c, even as $\rho_a(T)$ remains metallic. These observations call for a more careful investigation into microscopic sources of nematicity, especially in view of the resistivity anisotropy peaking (maximal nematic response, χ_N) around a doping where SC $T_c(x)$ is highest. Thus, the incoherent "normal" state gives way to either (a) a nematic phase (at least in 122-FePn), with a T-O distortion and subsequent SDW order, upon underdoping, or (b) to an unconventional SC with a dome in T_c versus x, or (c) to a renormalized Fermi liquid metal at low T upon overdoping.

These findings in 122-FePn conflict with *pure* spin fluctuation models ((i) and (ii) above), and, in addition to (i),(ii), in view of the insulator-like $\rho_b(T)$, weakly correlated models as well. Orbital-based models do identify the T-O distortion with multi-orbital (xz, yz) correlations [6], but nematicity has hitherto not been considered there. Finally, how such a coupled nematic-plus T-O transition could arise as an instability of the "high"-T "strange" metal [1, 7] remains open (the T-O transition generically precedes AF-spin density wave (SDW)). Here, we address these issues in detail using LDA+DMFT. Our philosophy is to identify the germs of the emergent order in the residual (two-particle) interactions present in the high-T incoherent fluid. Thereupon, extending our earlier LDA+DMFT calculations to a postulated orbital-nematic (ON) state, we show how the unusual features above find natural explication within a theoretical picture where an orbital nematic order arises as a direct instability of a sizably correlated bad metal.

In an incoherent metal, inter-band one-electron mixing is *irrelevant* in a renormalization-group (RG) sense, akin to the situation that occurs in weakly coupled $D = 1$ Luttinger liquids [8]. This inhibits a smooth crossover to a correlated FL, simultaneously favoring ordered states arising directly from the incoherent metal. The underlying reason is that when one-electron mixing is irrelevant, the corresponding second-order process, corresponding to two-particle intersite *and* inter-orbital hoppings, turns out to be the most relevant. In our earlier LDA+DMFT for the "normal" state, an incoherent metal arises from an Anderson orthogonality catastrophe-like effect due to orbital-selective (OS) Mott physics. The general two-body interaction, obtained at second order from a (now RG-irrelevant) one-electron inter-band term, $t_{ab} \sum_{<i,j>,\sigma}(c_{ia\sigma}^\dagger c_{jb\sigma} + h.c)$, is $H^{(2)} = \frac{1}{2} \sum_{a,b,k,k'} V_{ab}(k, k') c_{a,k,\uparrow}^\dagger c_{b,-k',\downarrow}^\dagger c_{b,-k',\downarrow} c_{a,k',\uparrow}$, where $a, b = xz, yz$ and the scattering vertex is $V_{ab}(k, k', \omega) = g^2 \chi_{ab}(k - k', \omega)$ with $\chi_{ab}(k, \omega)$ being the inter-orbital susceptibility. The static, nearest- and next-

nearest neighbor parts of $V_{ab}(k,k')$ are $V_{ab}^{(1)}(k,k',0) \simeq \frac{4t_{ab}^2}{U'+J_H} \simeq O(200-250)$ meV and $V_{ab}^{(2)} \simeq \frac{4t_{ab}'^2}{U'+J_H} \simeq O(120-160)$ meV, consistent with magnetic excitation energy scales from INS data. These *residual* interactions contain coupled, inter-orbital charge and spin fluctuations in particle-hole (p-h) *and* cooper (p-p) channels, immediately suggesting the possibility of both, mutually competing instabilities [9] from the "high" T incoherent metal. In earlier work, we showed how an unconventional SC with the right gap symmetry indeed arises from the cooper instability above [10]. Here, we study how a p-h decoupling of $H^{(2)}$ above in the situation specific to FePn leads to the ON instability. We decouple $H^{(2)}$ in the p-h sector as $H_{MF}^{(2)} = \sum_{a,b,k,\sigma}(\Delta_{ab}^{(n)}(k,k')c_{a,k,\sigma}^\dagger c_{b,k',\sigma} + a \rightarrow b)$, with $\Delta_{ab}^{(n)}(k,k') = \frac{1}{2}V_{ab}(k,k',0)\langle c_{a,k,\sigma}^\dagger c_{b,k',\sigma}\rangle$. Consistent with lattice symmetries, we write $V_{ab}(k,k') = \sum_l V_{ab}^l \eta_l(k)\eta_l(k')$, with $\eta_l(k)$ being the irreducible representations of the D_{4h} point group. Then it follows that $\Delta_{ab}(k \simeq k') = \sum_l \Delta_{ab}^l \eta_l(k)$, and, for the frustrated geometry of FePn, $\Delta_{ab}(k) = \Delta_{ab}^{(1)}(c_x+c_y)+\Delta_{ab}^{(2)}c_x c_y$ where $c_x = \cos k_x$, etc.

Interestingly, $N^z \simeq \langle n_{xz} - n_{yz}\rangle$ with $\langle N_a\rangle = \langle \sum_k \Delta_{ab}^{(n)}(k)c_{a,k,\sigma}^\dagger c_{a,k,\sigma}\rangle$ (with $a = xz, yz$), which is now finite, is exactly associated with *orbital* nematicity. And $\alpha = \Delta_{ab}^{(2)}/\Delta_{ab}^{(1)} \simeq 0.7$. A $\langle N\rangle = \frac{\langle n_{xz}\rangle - \langle n_{yz}\rangle}{\langle n_{xz}\rangle + \langle n_{yz}\rangle} \neq 0$ lifts the degeneracy of the xz, yz bands in the T-structure, as seen by considering that: (i) its form factor, $\Delta_{ab}^{(n)}(k)$, differs from those for the xz, yz bands (ii) in the real FePn structure, details of $p-d$ orbital overlaps along a and b axes leads to important differences in the xz, yz band dispersion [11] already at LDA and Hartree-Fock (HF) levels. With an anisotropic form factor in (i) above, we get $\langle N_e\rangle > 0$ [12], as in bilayer ruthenates, already at HF level. Note that this is now an ON instability arising from *an interplay between residual interactions* and *the renormalized LDA+DMFT band structure*, and arises directly from the incoherent metal. Our mechanism is thus radically different from one where ordered states arise from a weakly correlated LFL metal [4]. A finite $\Delta_{ab}^{(n)}(k)$ will split the xz, yz degeneracy above and modify the xz, yz band *dispersion* anisotropically, with an increase in xz orbital character in the "reconstructed" FS across the T-O transition, as noted recently [11], an effect we will consider in future work. The *symmetry* dictated coupling to the sizable magneto-elastic interaction in FePn will reinforce this splitting by coupling $\langle N\rangle$ linearly to the strain. This acts like a sizable "external" field, potentially enhances extant nematic order and will smear the nematic transition, making it impossible to separate nematicity from the T-O distortion.

If the xz band is thereby pushed below its position in the T-phase, it is not hard to deduce (see results below) that $n_{xz} > n_{yz}$, giving a finite value for $\langle N\rangle$. Strong local correlations (U, U', J_H) will now have a stronger ("Mott") localizing effect on the (more populated) xz band states, and we propose that the shortening of the lattice constant b is thus a direct fall-out of this correlation-induced instability to the ON phase. This will immediately induce the T-O distortion ($b < a$) and create conditions propitious for a $\mathbf{q} = (\pi, 0)$ AF instability. Enhancement of Mottness in the xz sector, together with the incoherent "normal" state above T_c, is also consistent with the observation of an insulator-like $\rho_b(T)$ below 200 K in the 122-systems, and cannot be found in weak-coupling pictures. Thus, the observed ON-phase (competing with U-SC) is intimately linked to *enhancement* of the OS incoherence, already a salient feature of the "normal" state. Sizable multi-orbital (MO) correlations and OS Mottness thus turn out to be *necessary* for even a qualitative reconciliation of these unusual features, as we detail below.

Extending earlier LDA+DMFT studies, we focus on a *four band* model, since the ON phase involves predominantly planar bands. Defining $\delta = \frac{b-a}{b+a}$ as the orthorhombicity, the orbital-lattice coupling is $H_{e,l} = \lambda\delta(n_{xz} - n_{yz}) + \frac{C_{s0}}{2}\delta^2$, *a la* the more familiar Jahn-Teller coupling in MO systems. This self-consistently renormalizes *both*, $\langle N\rangle$ and δ, as alluded to before. Since *ab initio* values of λ, C_{s0} are not known, we adopt the strategy of using trial values for $\langle N'\rangle = \langle n_{xz} - n_{yz}\rangle_0 = -\frac{C_{s0}\delta}{\lambda}$ in addition to the $\langle N_e\rangle$ computed from $\langle \sum_{k,\sigma}(n_{k,a,\sigma} - n_{k,b,\sigma})\rangle$ computed within LDA+DMFT [12]. We input trial values of $\langle N\rangle = \langle N_e\rangle + \langle N'\rangle$ as "external" symmetry breaking fields in the xz, yz orbital sector, and recompute the LDA+DMFT spectral functions in the sizably correlated multi-band Hubbard model [13], but now including the finite $\langle N\rangle$ above, along with $U = 4.0$ eV, $U' = 2.6$ eV and $J_H = 0.7$ eV, as used in earlier work. $\langle N^z\rangle$ is now self-consistently computed within DMFT from the converged $G_{xz}(k,\omega), G_{yz}(k,\omega)$.

We now discuss our results. In Fig. 1, we show the (converged) LDA+DMFT spectral functions for the in-plane d orbitals with $\langle N\rangle = 0$ and $\langle N\rangle \neq 0$. Clear low-energy OS incoherence is seen in the "normal" metallic (T) phase, a direct consequence of sizable MO correlations, as noticed by many workers [13]. With $\langle N\rangle \neq 0$, closer examination reveals interesting changes germane to the above discussion: (i) $\langle N\rangle > 0$ and "zeeman field" ($h_z > 0$) remove the $xz - yz$ degeneracy of the T-phase. With $U = 4.0$ eV, $U' = 2.6$ eV, MO Hartree shifts from sizable MO correlations renormalize $h_z = \delta$. DMFT values for $\langle N\rangle$ are found to be 0.049 for $x = 0.2(\delta = 0.4)$ eV and 0.0315 for $x = 0.4(\delta = 0.24)$ eV, vanishing in the T phase. Concomitantly, $\delta(T)$ follows the ON order parameter via $\delta = -\frac{\lambda}{C_{s0}}\langle N\rangle$. The dynamical self-energies $(\Sigma_a(\omega))$ from DMFT then drive sizable *changes* in spectral weight transfer (SWT) over energies $O(1.0-2.0)$ eV in response to the renormalization of δ. This feature is generic to strongly correlated quantum matter: observation of sizable SWT across the T-O transition should
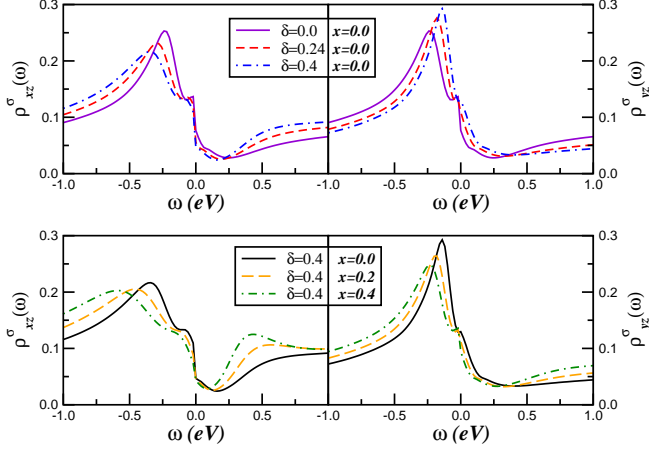
FIG. 1: (Color online) Orbital-resolved LDA+DMFT (with $U = 4.0$ eV, $U' = 2.6$ eV and $J_H = 0.7$ eV) density-of-states (DOS) for the Fe d orbitals for three δ values (top) and three doping values with fixed $\delta = 0.4$ (bottom). Large-scale and *anisotropic* dynamical spectral weight transfer accompanying the incoherent metal-nematic metal transition is clear, in good accord with optics [16].

FIG. 2: (Color online) Imaginary parts of the orbital-resolved LDA+DMFT self-energies for $\delta = 0.4$, showing sub-linear in-ω dependence for ω close to E_F, reflecting an incoherent metal. Also, the sharp pole-like feature in ImΣ_{xz} progressively sharpens and moves closer to E_F with δ, reflecting enhanced OS incoherence.

put our approach on firmer ground. (ii) revealingly, the pseudogap (PG), already a feature of the "normal" state, deepens for the xz band and *reduces* for the yz band. Concomitantly, the pole-like structure in Im$\Sigma_{xz}(\omega)$ progressively sharpens up and moves closer to E_F with increasing x (Fig. 2) for not-too-large x. Finally, given sizable U', interband "dynamical proximity" effects induce sizable spectral changes in the $xy, x^2 - y^2$ spectra as well. Thus, OS incoherence, already present in the "normal" state [13, 14], is further enhanced across the ON transition (iii) Using $n_a = (-1/\pi) \int_{-\infty}^{E_F} \text{Im} G_a(\omega) d\omega$, we find $n_{xz} > n_{yz}$, as advertized above. Thus, $\langle N \rangle > 0$ and, given the enhanced (Mott-like) localization in the xz sector, the lattice constant b now shrinks relative to its value $(b = a)$ in the T-phase: this is precisely the T-O distortion. With finite $\langle N \rangle \neq 0$ tied to enhanced Mott-like localization in the xz-sector, one can now write an *effective* ferro-nematic pseudospin model [6]: $H_{eff} \simeq -j \sum_{<i,j>} N_i^z N_j^z - \delta(T) \sum_i N_i^z$, and, with $\delta(T)$ as chosen above, *structural* anisotropy disappears at T_{T-O}. But strong precursor nematic correlations survive appreciably above T_{T-O} (cf. Ising model in a zeeman field).

If incipient nematicity exists above T_{T-O}, one expects electronic anisotropy to persist up to a sizable $T > T_{T-O}$: this should manifest as a in-plane resistivity anisotropy above T_{T-O}. In addition to the DMFT contributions, we also accounted for the influence of the (short-range) non-local ON (plus T-O) fluctuations on the *dc* resistivity (these are *necessary* close to T_{T-O}) by including
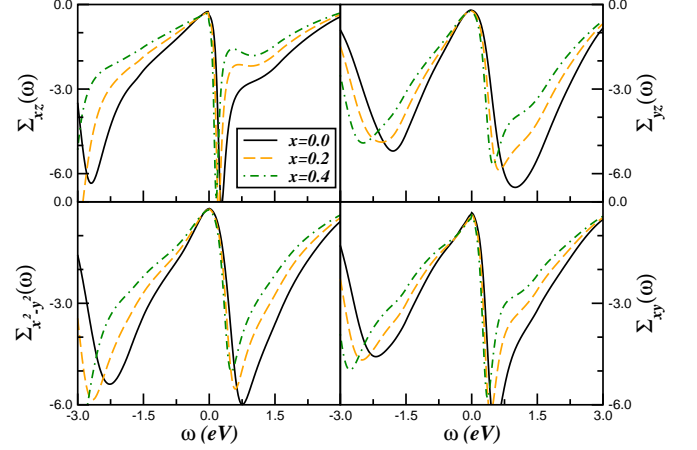
coupling of carriers to the *static*, T-dependent nematic susceptibility [15] using an embedded cluster-variation method for H_{eff} above. We used the DMFT effective masses and J_1, J_2 as above in the evaluation of the short-range order contribution to $\rho_{a,b}(T)$. This nicely fits within our DMFT-based view, since non-local (Ising) correlation effects are only included at a static level. In Fig. 3, we show the total *dc* resistivities $\rho_a(T)$ and $\rho_b(T)$ calculated within LDA+DMFT-plus static non-local ON correlations. Quite remarkably, we find that the in-plane resistivity anisotropy indeed survives up to $T \simeq 250 - 300$ K, sizably above $T_s = 150$ K, though it is a bit overestimated at high T. It is instructive to notice that this anisotropy already exists within DMFT (inset to Fig. 3), and is enhanced sizably near T_{T-O} due to non-local nematic correlations (which peak at T_{T-O} in our H_{eff}), in agreement with data. Here, $\rho_b(T) > \rho_a(T)$ in spite of $b < a$ ($\delta(T) > 0$) naturally follows from increased tendency to (Mott) localization in the xz band sector (corresponding to orbitals pointing along b) across the ON-plus T-O transition (see Fig. 1). Very satisfyingly, this anisotropy *increases* with x, as indeed observed in experiment (in a 122-FePn recently, but this also could be seen in 1111-FePn systems, where T_{T-O} and T_{SDW} are separated). We emphasize that this is in conflict with pure $J_1 - J_2$ *spin-only* models, where one expects $\rho_b(T) < \rho_a(T)$. It is also inconsistent with a weakly correlated Landau Fermi liquid (LFL) view for same reasons. In contrast, a ON-plus T-O transition within our OS-Mottness view fulfils *all* above experimen-
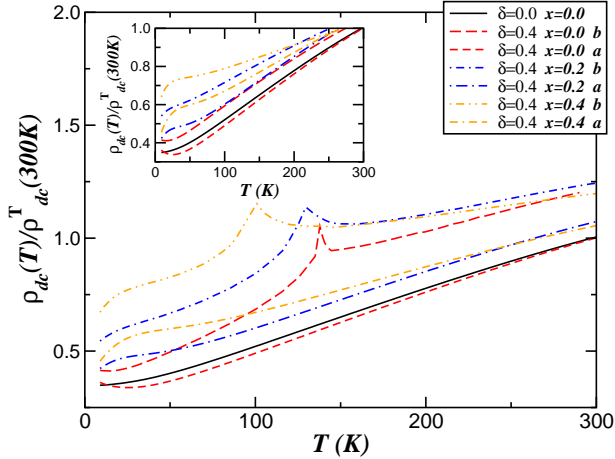
FIG. 3: (Color online) Resistivity versus temperature, showing the in-plane resistivity anisotropy with (main panel) and without (inset) the non-local, static (Ising) nematic correlations. Notice that DMFT already yields the correct sign of the anisotropy, and non-local corrections further enhance it near T_{T-O}, in good accord with data.

tal constraints [2]. Finally, the optical conductivity in DMFT (solely involving the full DMFT Green functions, $G_{xz}(\omega), G_{yz}(\omega)$), immediately implies different optical conductivities along a, b. Sizable SWT, $O(2.0)$ eV, across this ON-plus T-O transition in DMFT DOS above should show up as SWT in $\sigma_{a,b}(\omega)$ across T_{T-O}. Remarkably, both these features are indeed seen [16], lending strong support for our view.

A finite $\langle N \rangle > 0$ also implies ferro-orbital (FO) ordering, and, with sizable magnetoelastic coupling, leads to conditions propituous for the $\mathbf{Q} = (\pi, 0)$ striped SDW order to occur. Microscopically, the "localized" orbital pseudospins directly follow from enhanced tendency to Mott localization of the xz orbital states in our work. The "super-exchange" couplings in the effective $J_1 - J_2$ spin model, with "localized" spins arising from the incoherent Hubbard bands [18], are now explicit functions of the neighboring (Ising) orbital correlations along a and b [6], and, for realistic J_{1a}, J_{1b}, J_2, the striped AF-SDW state is indeed the ground state. Thus, AF-SDW order will always follow, or be coincident with, the T-O distortion, as is well known. Interestingly, the anisotropy of the spin-wave excitations [17] in $\mathbf{q}$-space also follows from such a "localized", frustrated model [18]. A weak coupling LFL approach [4] can also achieve this upon "tweaking" the ellipticity of the electron pockets phenomenologically. However, it is clearly inconsistent with $\rho_b(T) > \rho_a(T)$, as well as with the insulator-like $\rho_b(T)$ above T_{T-O}. Finally, our work is not inconsistent with LDA+DMFT works for the AF-SDW state [19] where coexisting quasi-itinerant and localized band states are in-

voked; however, it bares the intimate connection between the ON-plus T-O transition and subsequent AF-SDW in FePn.

Thus, our work offers a unified rationalization of various unusual features seen recently in underdoped 122-FePn. The observed in-plane resistive and optical anisotropies are understood as consequences of an electronic ON-plus orthorhombic order arising as a p-h instability (hence, competing with U-SC [9]) of the "high" T OS-incoherent metal. Finding of nanoscale nematic domains in 122 systems, as well as nanoscale electronic order in the 1111 systems is not inconsistent with effects of (doping induced) static disorder on our ON-plus orthorhombic state, which falls into the "liquid gas" universality class of an Ising model in a (random in presence of disorder) zeeman field [20]. To the extent that such fluctuating ON order will induce a spin gap and two distinct local "environments" (corresponding to $\langle N \rangle = +n, -n$), this should "rationalize" NMR data [3] as well. Our work opens up an intriguing possibility: an ON order with a T-O distortion, with subsequent stripe-AF, might be the primary competitor to unconventional SC in FePn, and establishes the important role of sizable MO correlations and OS Mott physics in this context.

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